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Torrefied, spherical biomass pellets through the use of experimental design

Andrew Duncan, Andrew Pollard *, Hachimi Fellouah

Department of Mechanical and Materials Engineering, Queen's University, Kingston, Ontario, Canada K7L 3N6

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ABSTRACT

Non-torrefied biomass pellets produced using extrusion methods are mechanically weak and therefore produce fines/dust during handling and transportation, readily absorb moisture, remain biologically active, have low specific energy content together with poor flow and handling characteristics. A new spherical, torrefied biomass pellet is introduced that uses a compression method. The resulting pellet addresses the above noted shortcomings of extruded pellets. Designs of Experiments were constructed to examine the responses of these new pellets. Tests of tensile strength, ultimate and proximate analysis, abrasion resistance, moisture sensitivity, and mass and dimensional characteristics are considered. Pellets were found to have tensile strengths between 855 and 2161 N, absorbed between 11.0 and 29.4 mass% water, but returned to near ambient conditions in less than 24 h, and had a range of gross calorific values between 17.5 and 21.1 MJ/kg and had 100% abrasion resistance. The Designs of Experiments were able to produce models with a 95% confidence level for torrefaction, tensile strength, gross calorific value, pellet size, density, and moisture absorption and evaporation. Torrefaction temperature was found to be a dominant influence on the outcome of most pellet characteristics, yet the process is non-optimised in this work and in general is not well understood.

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1. Introduction

Densification of biomass is important as it relates to its material properties. In its raw form, biomass is uneven, fluffy, dusty, and these characteristics make it difficult to store, transport and utilise [1]. Demirbas [2] noted that biomass properties are highly variable and that its calorific value is low in comparison to coal. It is evident that densification of biomass improves handling, reduces transportation costs and enhances bulk energy density [3]. Lehtikangas [4] noted that shorter pellets are easier to handle.

Although densification of biomass through pelletization improves some characteristics, problems remain so that they may be not well suited as a renewable energy source in for example power plants. Pellets produced by a typical ring die mould are weakly bound together, create dust and fines when transported and handled, may require a binder, and have a high affinity to moisture that leads to both physical degradation and possible microbial growth.

The storage of pellets often requires they be dropped through up to 10 or 15 m, [5]. Pellets may be pneumatically handled but this method often results in pellet breakage and the production of fines or dust. Kaliyan and Vance Morey [6] categorised damage types as due to: compression forces, impact forces and shear forces.

Thermal treatment (torrefaction) has been used as a method to create a hydrophobic material. Here, torrefied pellets will be referred to as "black" pellets, and non-torrefied, or raw pellets will be referred to as "white". Bergman et al. [8] explained that torrefaction is the thermal treatment of biomass in an oxidizer free environment at heating rates <50 °C/min to a temperatures between 250 °C and 280 °C. Torrefaction alters the molecular structure of the lignocellulosic components of the wood through a number of reactions that render the material more hydrophobic than non-torrefied material, Bergman et al. [8]. It also results in a more uniform product, which is a useful feature if pellets are used to displace fossil fuels in large scale combustion facilities. Black pellets were found to be stronger than white pellets and were more hydrophobic and able to withstand submersion in water without disintegrating, according to Bergman [9].

Pellet durability, particle size distribution and hygroscopic nature are most affected by material properties [5]. In addition, Lehtikangas [5] found that wood pellets typically tend to absorb moisture readily from the environment. White pellets are hygroscopic. The moisture in pellets lead to mechanical degradation, increased fungal and bacterial growth, which lead to increased material temperature, and in the worst case, the opportunity for spontaneous combustion.

The moisture content of white biomass pellets, or raw material requires that additional energy and consequently costs be bourne to dry it prior to combustion [4]. In addition, this moisture in the pellets can cause for higher costs in transportation as it effectively

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* Corresponding author.
E-mail addresses: andrewduncan@me.queensu.ca (A. Duncan), pollard@me.queensu.ca (A. Pollard), hachimi.fellouah@usherbrooke.ca (H. Fellouah).

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lowers the bulk energy density, which in turn raises the bulk price for the fuel.

The Gross Calorific Value (GCV) of white pellets is low compared to fossil fuels. Some light volatile compounds in biomass contribute to this lower GCV. These compounds may include, in varying quantities: acetic acid, water, methanol, formic acid, lactic acid, furfural, hydroxyl acetone, phenol, carbon dioxide, carbon monoxide, hydrogen and methane [7]. Torrefaction of biomass helps to devolatilize these compounds from the source material, which can decrease the overall mass through the loss of non-combustible compounds. The result is that the net energy density of the material increases on a mass basis. It is noted, however, that the physical and chemical properties of torrefied biomass depend highly on the torrefaction temperature and consequently it's potential to be used as solid fuel, see, for example Chen et al. [18].

The characteristics of biomass pellets are measured by standards organizations including CEN/TS, ASTM, and DIN. For fuel standards, the ultimate and proximate tests include elemental analysis, analysis of ash content, moisture content, gross calorific value, fixed carbon content, and volatile content through ASTM standards: D3302, D3174/5142, D5865, D4239, D5373, D3176. The corresponding CEN/TS standards are 14918, 14774, 15104, 15289, 15148. In addition to these tests, mechanical and chemical characteristics of fuel pellets which include fuel additives, binders, extraneous material, fuel particle size and dimension, mechanical durability, fines, fractional size distribution of raw material, bulk density, specific density, and angle of repose are measured through CEN/TS standards: 14961, 15210–1, 15149–2, 15149–3, 15103, and IMO 260E [10]. These standards are for the main part similar, and test for the qualities of wood pellets for their use as an energy fuel. These tests incorporate durability through mechanical tumbling of pellets, sieve shakers to determine fines content and volumetric packing to determine bulk density for example.

Current cylindrical-shaped white pellets are not well suited as a fuel as they cause storage, handling and transportation (SHT) issues. The costs associated with the transportation and processing of white wood pellets are responsible for a great deal of the difference between using biomass as a fuel rather than coal, Uslu et al. [11] and Mahmudi and Flynn [12]. To assist to displace coal, an ideal pellet should address problems with the SHT issues. A pellet made from biomass that could directly replace coal with minimal impact on existing infrastructure while manufactured and supplied at a cost equivalent to coal would be the ideal solution.

To address the above noted problems with either white or black pellets, a new fuel pellet has been invented (Provisional patent US 61/441,510). The densification method is also a novel technology. The torrefied pellet is spherical and contains no binders. It is amenable to be transported in a hydraulic or pneumatic pipeline and the surface area to volume ratio is minimal.

To study the characteristics of this new fuel pellet, three Design Of Experiments (DOE) were created. The DOEs were three-factor, two-level full factorial designs with inputs of Material Moisture Content (MC), Material Particle Size Distribution (PS), and Torrefaction temperature (T). The DOEs were created using the responses from a range of physical and chemical tests to produce linear models to enable for the prediction of pellet characteristics in any subsequent tests. Since no work had been done on the creation and testing of spherical biomass pellets, the DOEs were limited to screening designs to extract a basic understanding of the mechanisms affecting their physical and chemical characteristics. The physical and chemical tests that yielded responses included crush tests for tensile strength, abrasion tests via a tumbling can method, ultimate and short proximate analysis, moisture sensitivity, measurements of size and density, and the effective level of torrefaction applied to the biomass.

2. Methods and materials

The pellets consisted of 100% hybrid poplar supplied in chipped form from the Eastern Ontario Model Forest. This material was hammer milled, dried, sifted and sorted for size distribution and then reconditioned to the desired moisture content. Material was carefully weighed and then subjected to torrefaction and compression processes.

2.1. Material factor inputs for DOEs

The factor inputs, represented by the variables A, B and C in the DOE design were:

2.1.1. Particle size distribution (Factor input "A")

Particle sizes were divided using the Tyler mesh system. The sizes of material used were from Tyler Mesh numbers 14 (1.200 mm), 20 (0.853 mm), 28 (0.599 mm), and <28 mesh (<0.599 mm). The smallest particle size consisted of 40 mass% content of 14 mesh particles, and 60 mass% content of 20 mesh particles. The largest level particle size was comprised of 40 mass% content of size 28 mesh particles and 60 mass% content of <28 mesh particles. The particle size choice and factor level definition is counterintuitive and the reader when analysing these data should take care.

2.1.2. Material moisture content (Factor input "B")

The moisture content of the preconditioned material was used in the experiments. The low level of moisture was approximately 8% and the high level was approximately 15%. These values were chosen as they were found from previous work as an acceptable range over which densification of biomass into pellets could be performed successfully.

2.1.3. Torrefaction temperature (Factor input "C")

The torrefaction temperature was used as the final factor input. The lower temperature was 250 °C and the higher was 280 °C. These temperatures provided two different levels of torrefaction to identify how these torrefaction temperatures were affected by the moisture content and particle size.

2.2. Test procedures

The responses that were used in the DOE creation were obtained by the results of physical and chemical tests and these are explained in the following section.

2.2.1. Tensile strength

A calibrated 9072 kg (20,000 lb) load cell was used to measure the force applied to the pellet. Pellets, placed in an Instron press, were oriented in a vertical fashion and data were acquired as the press applied force onto the pellets until failure occured.

2.2.2. Abrasion resistance

The pellets were placed in a tumbling can with 20 glass marbles. These marbles had an average mass of 5.2840 g and diameter of 15.86 mm, which is about the diameter of the spherical pellet ($d = 12.5$ mm). Each pellet was tested on an individual basis in the abrasion test. The marbles and pellets filled less than ¼ of the volume of the tumbler. The can was rotated at 60RPM for 10 min, which is 10RPM faster than ASABE standard S269.4 as referenced by Kaliyan and Vance Morey [6]. The method differed from the standard as the tumbling container was cylindrical and the abrasive material (the marbles) was different from the pellet material. It is not uncommon to add in more abrasive material to test very durable pellets. This is common in testing of feed pellets where hexagonal bolts have been included to test products [6].

2.2.3. Ultimate and short proximate analyses

Samples of the pellets that were destroyed in the crush test were sent to SGS North America, Minerals Analysis Division, Denver, Colorado. The samples were analysed for moisture, ash, gross calorific value, sulphur, nitrogen, and a full ultimate analysis.

Test methods and relative accuracies for the ultimate and proximate tests were as follow (note 1BTU/lb = 2326 J/kg) (see Table 1):

2.2.4. Size, shape and density

Pellets were measured using Vernier callipers (±0.02 mm) and weight to within 0.0001 g and a repeatability of ±0.0001 g. The density was simply calculated using these data.

2.2.5. Moisture sensitivity

2.2.5.1. Dimensional measurement. Pellet dimensions prior to the submersion test were done using digital Vernier callipers. When the pellets were removed from the water, they were re-measured. They were also measured at the beginning and end of the evaporation phase.

2.2.5.2. Mass measurement. Pellets were submerged in water at ambient temperature and pressure. The pellets were removed from the water and quickly weighed at measured intervals of time; the surfaces were pat dry with a paper towel prior to the mass measurements. The same analytical balance was used in the evaporation stage; however, the pellets were not dried at each measurement as there was no residual film or droplets of water on their surface.

2.2.6. Torrefaction

The degree to which material was torrefied was calculated based on the original and final pellet mass. The original mass did contain material moisture of approximately 8%, 11.5% and 15% depending on the particular experiment. These measurements were made on material at room temperature.

Table 1
Ultimate & proximate analysis test statistics, methods, and relative accuracy. Provided by SGS North America. 1BTU/lb ~ 2324 J/kg.

Parameter	Test	Repeatability estimates
% Original moisture	ASTM D3302	+/−0.4%
% Ash	ASTM D3174/5142	+/−0.5%
GCV (BTU)	ASTM D5865	+/−150 BTU/lb
%S	ASTM D4239	+/−0.1%
%C	ASTM D5373	+/−1.0%
%H	ASTM D5373	+/−0.2%
%N	ASTM D5373	+/−0.1%
%O	ASTM D3176	–

3. Theory

3.1. Experiment formulation

The analysis by DOE is designed to provide an objective assessment of factor influence. Each DOE was used to produce a model that enables the prediction of the responses from a repeat experiment with a linear trend.

The DOE constructed was a simple screening design. This means that each set of experiments was a sole set of experiments, and there were no replicates of test points in each case. There were however, measurements of some common responses of mass, density, size, and torrefaction level in each of the three experiments.

Three destructive tests were performed on pellets: the tensile strength, abrasion and the water immersion tests. A repeat set of tests could not be performed on the products of a prior test. Therefore, three individual sets of identical pellets were formed to examine the characteristics of the pellets.

The set of pellets made for the second DOE were also used in the third DOE, as the second DOE did not physically alter the pellets in any measurable way. The test may in fact be more realistic as any pellets that are to resist moisture, must have first been transported to their destination, and would have experienced conditions and handling similar to the tumbling can test.

Only first-level and main interactions were examined in the DOEs, which is standard in a screening design where the predicted response over the region of interest is expected to be linear. To check for error and linearity, centre points were used.

Table 2 below displays one of the structured DOEs with the high and low values of the factor inputs being used to create the experiment. The treatment combination is used to explain the level of a particular factor in that experiment. The upper case letter refers to the factor, and the smaller case letter refers to those factors used in each treatment combination. The corresponding letters and factors are defined as “A” which is particle size distribution, “B” being moisture content, and “C” being torrefaction temperature. To minimise bias error in the analysis of the results, the sets of pellets were created in a completely randomized manner.

A brief review of results from the experiments made it clear that there had been an error in the selection of the central values of the experiment. The screens used to fraction the material particle size were not of a linear scale, and therefore, the low, central and high values of the particle size did not fit the hypothesis that the responses should be linear. This meant that the centre point runs could not be used to check for linearity of the responses, and the central points needed to be removed.

3.2. Data analysis

JMP, a statistical analysis program, was used to perform calculations to create the prediction models. The responses that were observed for each of the tests formed the solution to a system of linear equations. These linear equations were solved to determine the coefficients required to produce a model of the form:

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_4 x_1 x_2 + \beta_5 x_1 x_3 + \beta_6 x_2 x_3 + \varepsilon \qquad (1)$$

Table 2
Treatment combinations for a 2^3 DOE.

Run	A	B	C	Treatment combination	Run	A	B	C	Treatment combination
1	–	–	–	1	5	–	–	+	*c*
2	+	–	–	*a*	6	+	–	+	*ac*
3	–	+	–	*b*	7	–	+	+	*bc*
4	+	+	–	*ab*	8	+	+	+	*abc*

where “y” is the predicted model response, “β_n” is the particular model coefficient, “x_n” is the factor input level, and “ε” is the remaining error in the model approximation.

3.2.1. Visual examination

The responses of the experiments were plotted against the input factors using multivariate plots. These plots indicate what main or interaction effects may be of interest in the creation of a linear model. The model was then plotted against the actual response to assess the validity of the model. Main and interaction effects were then plotted. If there were interaction effects between factors, these effects would be of significant interest to include in the refined model. The slope of the line representing the interaction effect also indicated the relative degree of impact of that factor and whether it influenced the response either negatively or positively. These plots must be treated holistically as a guide in refining the model.

3.2.2. Calculated data

Sample calculations were done in Excel that verified the accuracy of the outputs produced in JMP. Confidence was needed that the model fit these data well, and that the model actually represented the true physical and chemical behaviours of the tests samples. This was achieved through the examination of the coefficient of determination, R^2, R^2_{adj}, see [14], confidence intervals of linear fit, F_o values of the model (see [14]), and the appropriateness of including a model coefficient (see [14]) t_o values of each parameter and their significance. The F_o and t_o value for each of the models was restrained by a 95% confidence interval as a cut off for inclusion. For a more detailed background of the statistical analysis used in this work the reader is referred to [14].

3.2.3. Residuals

Two residual error plots between the model and the experiment were generated: one against the row number of the experiment to determine bias, and the second against the predicted model to check for normal distribution. Experiments were performed in a random order to eliminate bias. The residuals were examined for patterns that may suggest underlying factor effects that had not been included in the model.

The steps above were performed on the initial model output from JMP. This initial model was comprised of all main and interaction effects. To refine the model, the statistics described were used to determine which factors to retain in the model. By constraining the F_o and t_o values to a 95% confidence level, the less accurate terms in the model were eliminated, and the model was refined to the best available description of the system.

4. Presentation and discussion of results

4.1. Physical and chemical test results

Table 3 provides the results that were used to create the models. Some data was found to be insufficient to be able to produce a model with a 95% confidence interval, and were therefore excluded. The highlighted rows identify the results that were used to form models in DOE 1 and the remaining non-highlighted rows were used for DOE 3. The results obtained from DOE 2 did not produce any models and are therefore excluded.

4.1.1. Load

Tensile strength was calculated based upon the ranges of load the pellets could withstand. A point load test on the spherical pellets was used:

$$\sigma_f = \frac{F}{\pi r^2} \tag{2}$$

where F is the force applied by through the platens and r is the radius of the pellet, describes the tensile strength of pellets.

With the minimum and maximum loads substituted into (2), spherical pellets are found to have tensile strengths between 3.38–17.06 MPa. These results would ideally be compared to the tensile strength of torrefied cylindrical pellets, but no torrefied pellets were commercially available at the time of this work. The range over which the pellet strength can be adjusted through the alteration of the factor inputs suggests that the pellets can be designed to be very strong to resist damage, or designed to be weak to minimise the energy requirements from the use of a pulveriser at a power plant.

4.1.2. Gross Calorific Value (GCV)

Torrefaction increases the GCV of the material and can increase the energy per unit volume. The GCV was altered by torrefaction but did not always result in an increase, as was hypothesised. The original GCV of “white” poplar was determined to be 17.92 ± 0.35 MJ/kg from the tests performed by SGS North America. The torrefied spherical pellet material were analysed as well by SGS, and were found to be 17.55 MJ/kg < GCV < 21.13 MJ/kg.

The torrefaction process did not always increase the GCV of the pellets. The energy balance for the torrefaction reaction should be examined to help identify possible reasons for this outcome. The

Table 3
Responses of physical and chemical results for DOE 1 and 3.

	1	*c*	*b*	*bc*	*a*	*ac*	*ab*	*abc*
Load (N)	2161.23	855.17	2089.37	1091.15	1863.85	1113.05	1674.61	1378.08
GCV (MJ/kg)	17.82	20.05	17.55	17.56	21.13	20.17	18.14	18.08
Moisture gain								
(% Size change)	15.00	7.78	17.07	7.74	17.93	5.89	16.24	5.08
(% Mass increase)	24.33	15.61	29.37	17.58	26.21	11.03	27.45	11.34
Moisture evaporation								
(% Size change)	8.05	3.75	9.22	2.98	10.36	2.80	9.04	2.43
Density (kg/m^3)								
DOE 1	1354.00	1331.00	1281.00	1371.00	1371.00	1368.00	1343.00	1300.00
DOE 3	1316.93	1334.18	1324.11	1318.25	1365.98	1338.42	1320.40	1301.68
Pellet size								
(mm)	13.67	13.88	13.88	14.09	13.61	13.58	13.61	13.57
Torrefaction								
Residual mass fraction DOE 1	0.83	0.74	0.81	0.73	0.84	0.73	0.81	0.72
Residual mass fraction DOE 3	0.82	0.74	0.81	0.72	0.83	0.74	0.79	0.71

loss of energy in this example demonstrates that the process did not optimise the densified energy content.

4.1.3. Moisture sensitivity

The hydrophobic feature of the spherical pellets was very high; however, they did absorb water when immersed in a water bath. The main difference, though, was that the pellets shed 95% of the water that they had absorbed within 13 h of removal from the water bath. When placed in ambient conditions the pellets absorbed on average 3% moisture.

Pellets created in this work were of higher quality than those examined in similar studies. Pellets created in the TOP process at Energy Centre of the Netherlands (ECN) were found to survive a submersion test and maintain their structural integrity; however, some pellets did disintegrate [9]. Additionally, Hartley and Wood [15] tested the hygroscopic nature of pellets in a controlled humid environment and found that when exposed to "between 93% and 100% humidity", the pellets disintegrated. Both papers concluded that torrefaction helped to make the pellets hydrophobic.

4.1.4. Mass, density & shape

Pellets created for and analysed in this work have densities in excess of those described in the literature. The shape of the spherical pellets aids transportation and handling. The shape reflects the opportunity to maximise packing [16].

4.1.5. Torrefaction

The torrefaction of the material was a crucial thermal treatment that produced the majority of the characteristics of the pellets. These include conversion from hydrophilic to hydrophobic, increased specific energy content, and pellet homogeneity through devolatilization. The results show that the torrefaction produces expected mass losses associated with the devolatilization of the material. However, as the rest of the responses indicate, torrefaction is a much more complex process that alters the physical and chemical properties of the pellets. Thus, it is of more interest to examine the results of torrefaction as they pertain to the models formed from the DOEs.

4.1.6. Abrasion resistance

The abrasion resistance of the pellets was very high, and there was no visible sign of dust in the tumbling can or damage to any of the pellets or marbles. The typical test of screening for fines did not apply here, as there were no fines. After weighing the pellets to determine if there was any mass loss, it was found that the pellets were all within ±0.15% of their original mass before they were placed in the tester. The fact that some pellets gained mass suggests that the test could not accurately account for any definitive mass loss in the pellets. It is speculated that the mass changes was more likely due to moisture absorption from the atmosphere. The fact that no fines were produced suggests that these pellets are 100% abrasion resistant, which would classify them as the most durable pellets created without the use of a binder.

4.2. Model summaries

The responses have been listed in order of importance to the prediction of response based on their weighting (based on 95% confidence level). As a review, the X1, X2 and X3 terms refer to the particular factor inputs of Particle Size Distribution (**PS**), Material Moisture Content (**MC**), and Torrefaction temperature (**T**), respectively.

4.2.1. DOE 1

Table 4 provides the results from DOE 1 and the model coefficients that were determined to be able to represent the responses at a 95% confidence interval.

4.2.1.1. Load (−X3, +X1X3, +X2X3). The sustained load applied to the pellets were most affected by **T**. Torrefaction increases the friability or lowers the toughness, or ability to resist fracture when stressed. Although the model suggests that increased temperature lowers the tensile strength, it must also be noted that the interaction of **PS**&**T** and **MC**&**T** increase the tensile strength. It is speculated that the smaller the **PS** the higher the sustainable load due to increased number of surface bonding sites. The **MC** may have allowed for more strength due to the delay of the torrefaction reaction, which lowers the strength.

4.2.1.2. Torrefaction (−X3, −X2, −X1X3, −X1X2, +X2X3). The torrefaction temperature, T, is directly related to the mass loss and the higher **MC** means that more material was removed during the torrefaction process. It is speculated that the water acted as a catalyst to enable more material to be devolatilized. Both **T** & **PS** and **MC** & **PS** also increase mass loss. Smaller particles provide increased surface area thereby increasing the mass loss through their surfaces. The increased temperature may also accelerate mass loss from smaller particles. The combined effect of increased **MC** & **T** is speculated to be due to diversion of energy to evaporate moisture that would otherwise be used in the devolatilization process. This would slow down the reactions, and over the same time, decrease the overall mass loss. It is recommended that additional experiments be performed to investigate this effect.

4.2.1.3. GCV (−X2). The model for GCV immediately appears questionable as **T** was expected to be the main input to increase the value. The model for the GCV was included in the results as it had an F_o and t_o of at least 95%. The results appear not to follow a linear trend based upon **T**, so that the hypothesis that this response should be linear may be invalid. The **T** input maybe optimised to maximise GCV at some temperature intermediate to the values used in this work. Additionally the torrefaction process itself may be responsible for the outcome of the GCV, and should be examined. If the model is valid however, the interesting observation is that **T** seems to be not as important as the **MC**. It is speculated that **MC** may act as a catalyst to allow more volatiles to be driven off at lower temperatures and these volatiles may have been comprised of high calorific value constituents.

Table 4
DOE 1 model coefficients.

DOE 1	Intercept (β0)	X1 (β1)	X2 (β2)	X3 (β3)	X1X2 (β4)	X1X3 (β5)	X2X3 (β6)
Load (N)	1528.4567	0	0	−417.1758	0	158.8867	97.3774
Torrefaction (% material remaining)	77.6311	0	−0.8932	−4.8583	−0.2311	−0.2912	0.2081
Size (mm)	0	0	0	0	0	0	0
Density (kg/m^3)	0	0	0	0	0	0	0
GCV (MJ/kg)	19.1515	0	−1.0000	0	0	0	0

Table 5
DOE 3 model coefficients.

DOE 3	Intercept ($\beta 0$)	X1 ($\beta 1$)	X2 ($\beta 2$)	X3 ($\beta 3$)	X1X2 ($\beta 4$)	X1X3 ($\beta 5$)	X2X3 ($\beta 6$)
Torrefaction (% material remaining)	77.0173	0	−1.3582	−4.4282	−0.6060	0	0
Size (mm)	13.7361	−0.1437	0.0513	0.0437	−0.0539	−0.0612	0
Density (kg/m^3)	1327.1017	0	−11.9689	0	−9.7791	−7.2064	0
Mass absorption (% change)	20.3863	−1.3373	1.1136	−6.4764	0	0	0
Size absorption (% change)	11.5913	0	0	−4.9681	0	0	0
Mass evaporation (% change)	0	0	0	0	0	0	0
Size evaporation (% change)	6.0779	0	0	−3.0886	0	0	0

4.2.2. DOE 2

There were no models that were formed from DOE 2 as the response from the abrasion test did not produce useful data.

4.2.3. DOE 3

Table 5 provides the results from DOE 3 and the model coefficients that were determined that represent responses to a confidence interval of 95%.

4.2.3.1. Torrefaction (−X3, −X2, −X1X2). The **T** and **MC** inputs show the same characteristics in this model as in DOE 1for the same reasons. This is also true for the interaction of **MC** & **PS**. Two additional terms (−X1X3 and +X2X3) that were in the first model were not included here because they did not meet the 95% cut-off. Indeed, it is possible that they should not appear in the first model. As observed, they have the least magnitude of influence on the model.

4.2.3.2. Pellet Size (−X1, −X1X3, −X1X2, X2, X3). A low **PS** value, relating to the use of a larger particle size distribution indicated the size of the pellet would decrease. This may be due to increased bending and deformation of larger particles. The voids between these would not have filled as easily as if smaller particles were available to fill the gaps. A higher **MC** may have delayed the torrefaction reaction reducing the amount of material that was devolatilized, and subsequently leave more material in the mould. The **T** increased the pellet size, however this as the least significant term in the model does not represent how the system should perform, and may be an erroneous term.

4.2.3.3. Density(−X2, −X1X2, −X1X3). The **MC** is mainly responsible to lower the density of the pellets. This may be due to moisture loss, which could precipitate the development of increased numbers of porous spaces during devolatilization. The interaction of **PS** & **MC** and **PS** & **T** had a low confidence level and as in DOE 2, no model could be determined for density. Thus, the density may not represent the system well.

4.2.3.4. Massabsorption (−X3, −X1, X2). The temperature is the most important factor to increase hydrophobicity. This was observed through the decrease of mass of moisture absorbed as a function of **T**. Additionally, a higher **PS** also decreased mass absorption. It is thought that, as more particle surfaces bind together to produce better packing the overall pellet was also more hydrophobic. These results may also be partially explained through reduced capillary effect through better particle bonding. The **MC** may have delayed the torrefaction reaction, or may also have allowed for the venting of condensable hydrophobic volatiles, that would have increased the hydrophobic qualities of the pellets.

4.2.3.5. Size absorption (−X3). This response is correlated to mass absorption, as water is incompressible. The **T** was seen to decrease the mass absorption and therefore the pellet did not swell when torrefied.

4.2.3.6. Size evaporation (−X3). The pellets shrank in size in comparison to their swollen state after they were removed from the water and dried. **T** was found to hold the particles together and may have made the pellets more resistant to swelling. It is speculated that the pellets that were more torrefied (that is, higher **T**) had more elastic properties that allowed them to return to their original air-dried shape more effectively than pellets produced at lower **T**.

4.3. DOE factor input review

The results of torrefaction between DOE 1 and 3 were very similar. DOE 1 predicted that **T**, **MC**, and **PS** & **T** interaction were the most important effects to predict the torrefaction level. DOE 3 predicted that **T**, **MC**, and **PS** & **MC** were the dominant contributing factors. Both of these models produced very similar results. Interestingly, a higher torrefaction temperature does not necessarily translate into higher GCV. In these experiments, the off-gasses from de-volatilisation were not collected; therefore, the energy balance of the system is incomplete. The model for GCV appears in accord with this assessment as it was not determined to be based on torrefaction temperature, and thus may not actually produce a linear response.

Pellets torrefied at a higher temperature were weaker in tension and more friable which is in general agreement with the literature. The pellets were more hydrophobic than non-torrefied wood. As a result of the torrefaction, the pellets remained intact when submerged in water. Furthermore, the current study is the first to note that torrefied material readily sheds moisture. In addition to the hydrophobic, friable characteristics, these pellets were very resilient and resisted swelling and cracking due to water absorption and returned to their original state when removed from water.

The evidence from the responses indicate that pellets made from smaller particles are better than those made from larger particle size distribution, yet neither of these inputs were optimised to produce improvements. A smaller particle size distribution appears more preferable to increase the hydrophobic nature of the pellets.

The screening design may also be used as a starting point for the construction of an optimisation for pellet design. The optimisation could be used to design pellets for very particular applications.

5. Conclusion

The screening design provided data for an analysis of physical and chemical tests of the pellets in an objective way. The use of a DOE allowed an error and uncertainty analysis to be applied to the results of the tests. It was also able to produce a model that provided a basic understanding of the mechanisms that are required for the creation of pellets with particular characteristics.

DOE 1 showed that the load, the mass lost due to torrefaction, and the GCV are predictable to at least a 95% confidence level for this set of operating conditions. The load that the pellets could withstand was between 855.17 and 2161.23 N, which is a significant increase relative to known binder-less pellets. The GCV of the pellets was determined to be between 17.55 and 21.13 MJ/kg. DOE 2 was unable to produce any models, but from the observations, it is clear that the pellets are very durable and produce no fines. DOE 3 agreed with DOE 1 that torrefaction can be confidently predicted at a 95% level. DOE 3 was also able to produce models that predicted pellet size, density, mass absorption, size absorption, and size evaporation at greater than 95% confidence. Pellets absorbed between 11.03 and 29.37 mass% water when immersed in water for 24 h, yet readily shed the water when returned to ambient conditions. Their final dimensions differed between 2.43% and 10.36% from their original size.

The one factor that seemed to dominate all others is the torrefaction temperature. As torrefaction is not a well-understood treatment, Lipinsky et al. [17], it is clear that additional work in this area of biomass research is required.

The DOEs used single replicates, and confidence in the results of the experiments and the predictions by models should be treated accordingly. The use of centre points was unsuccessful due to an error in the preparation of the experiments. This does have consequences for the robustness of the model, and may help to explain the GCV data. Replicates of each test point should also be included to allow for a more definite set of results.

Finally, a stricter torrefaction process may be used to identify exactly how the process affects each of the responses. In addition, the DOEs here may be used as the basis of an optimisation of the pellets to meet end users needs.

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